Electronic Appendix A: Supplementary material to accompany the manuscript, "$Fe^{3+}/\sum Fe$ in Mariana Arc basalts and primary fO_2".

Screening for olivine interference in Fe-µ-XANES spectra

When collecting Fe-µ-XANES spectra on olivine-hosted melt inclusions, it is important to avoid hitting the olivine crystal with the beam during analysis. Olivine contains several weight percent of Fe^{2+} and even a very small amount of olivine interference will "contaminate" the pre-edge structure of Fe-µ-XANES spectra collected for melt inclusions and bias the result towards more reduced values. The region of XANES spectra at higher energies than the Fe-Kα absorption edge contains information related to Fe-coordination and can be used to distinguish glass structure (random and on average, uncoordinated) from olivine signal (strong coordination, Fig. A1). All melt inclusion and seafloor glass spectra were visually inspected and compared to spectra taken on San Carlos olivine and standard glasses from Cottrell *et al.* (2009) in order to screen for crystal contamination. Any spectra demonstrating signs of spectral features similar to those observed in San Carlos olivine were not considered in this study and additional spectra were collected to accommodate for this elimination.

Model liquid lines of descent

To constrain the effects of fractional crystallization on magmatic $Fe^{3+}/\sum Fe$ ratios, model liquid lines of descent that match the observed major element variations were generated using PetroLog3 (Danyushevsky & Plechov, 2011). The mineral-melt models that most closely replicate the natural data were chosen for each location, resulting in some variation in the models used from volcano to volcano. Individual model parameters

chosen to generate each LLD are provided in the figure captions to Fig A2-A6. If crystal fractionation is the main control on magmatic Fe redox, each model is meant to assess the extent to which Fe redox ratios may vary as a result of the relative incompatibility of Fe^{3+} in each fractionating phase. Implicit in this assessment is the assumption that fractional crystallization proceeds in a system closed to oxygen. Evidence from a global study of MORB glass suggests that closed system fractionation of olivine ± plagioclase ± clinopyroxene explains variations in $Fe^{3+}/\sum Fe$ ratios (Cottrell & Kelley, 2011). In our present study, each modeled LLD assumes a $D^{Fe3+} = 0$ and a D^{Fe2+} that varies according to the mineral-melt models chosen, for olivine ± clinopyroxene ± plagioclase fractionation. Magnetite mineral-melt models use a non-zero D^{Fe3+} that depend upon the composition of the melt. Each model was generated using a starting composition from a measured sample in this study.

Agrigan

Agrigan tephra samples Agri7, Agri04, and Agri05 all contain crystals of olivine, plagioclase, clinopyroxene, and magnetite. The clasts are red-brown, rounded and slightly weathered. In tephra sample Agri04, there are occasional clay fragments present. In tephra sample Agri05, there are occasional clasts of country rock that are red in color. The largest size fraction observed in these tephra samples is ~0.5 cm in diameter.

The modeled LLD begins with sample Agri-04-05 as the parental composition. The model crystallizes olivine isobarically at 1 kbar, then saturates clinopyroxene at 4.3 wt% MgO and then saturates plagioclase at 4.2 wt% MgO (Fig. 3a-d, A2). The whole rock and melt inclusion data for Agrigan from the literature and this study are broadly consistent with this model and in the case of all major elements except for FeO^{actual} and

$Fe_2O_3^{actual}$, the fractionation slope defined by the modeled LLD between 5-7 wt% MgO was used to correct for the effects of fractional crystallization (Electronic Appendix E). For FeO^{actual} and $Fe_2O_3^{actual}$, an empirical fractionation slope constrained by the trend in the melt inclusion data from this study was used (Electronic Appendix E).

Pagan

Tephra sample Paga8 is black in color and fresh. The largest size fraction observed in this tephra sample is ~1 cm in diameter. This tephra sample contains plagioclase, olivine, and clinopyroxene. Submarine pillow basalts are vesicular and have a glassy pillow rind. Pillow interiors contain plagioclase, clinopyroxene, and olivine.

The modeled LLD begins with sample HDP1147-R06 as the parental composition. Starting at 2.5 kbar and decompressing at a constant rate of 5 bar per 1ºC, the model crystallizes olivine and clinopyroxene, and then saturates plagioclase at 7.0 wt% MgO, and then magnetite at 6.5 wt% MgO (Fig. 3a-d, A3). The whole rock and melt inclusion data for Pagan from the literature and this study are broadly consistent with this model, except for TiO_2 and SiO_2 (Fig. A3). This may result from magnetite saturation too early or in too great abundance in the modeled LLD. It is also important to note that whole rock data may contain accumulated crystals that pull the whole rock compositions away from the true liquid compositions. Systematic offsets between melt inclusions and whole rock compositions in Al_2O_3 or CaO/Al_2O_3 variations with MgO are evidence for this phenomenon. In the case of all major elements except for TiO_2, SiO_2, FeO^{actual} and $Fe_2O_3^{actual}$, the fractionation slope defined by the modeled LLD between 5-7 wt% MgO was used to correct for the effects of fractional crystallization (Electronic Appendix E). For TiO_2, SiO_2, FeO^{actual} and $Fe_2O_3^{actual}$, an empirical fractionation slope constrained by

the trend in the melt inclusion and submarine glass data from this study was used (blue line on TiO_2 panel, Fig. A3; Electronic Appendix E).

Alamagan

Tephra samples Ala02 and Ala03 both contain olivine, plagioclase, clinopyroxene, and magnetite. Both tephra samples have vesiculated tephra fragments. The largest size fraction in Ala03 is ~2 cm in diameter, and ~1 cm in diameter in Ala02. Tephra sample Ala03 is altered, brown-red in color. Tephra sample Ala02 is black-brown in color.

The modeled LLD begins with sample Ala02-01 as the parental composition. Starting at 2 kbar and decompressing at a constant rate of 15 bar/ºC, this model crystallizes olivine, then saturates clinopyroxene at 4.6 wt% MgO and then saturates plagioclase at 4.5 wt% MgO (Fig. 3a-d, A4). The whole rock and melt inclusion data for Alamagan from the literature and this study are broadly consistent with this model and in the case of all major elements except for FeO^{actual} and $Fe_2O_3{}^{actual}$, the fractionation slope defined by the modeled LLD between 5-7 wt% MgO was used to correct for the effects of fractional crystallization (Electronic Appendix E). For FeO^{actual} and $Fe_2O_3{}^{actual}$, an empirical fractionation slope constrained by the trend in the melt inclusion data from this study was used (Electronic Appendix E).

Guguan

Tephra samples Gug11 and Gug23-02 are black in color and are fresh. The largest size fraction in both tephras is ~0.5 cm in diameter. Both tephra samples contain crystals of olivine, plagioclase, and clinopyroxene.

The modeled LLD begins with sample SD46-1-1 as the parental composition (Stern *et al.*, 2006). Starting at 2 kbar and decompressing at a constant rate of 5 bar/ºC, this model crystallizes olivine, then saturates clinopyroxene at 6.3 wt% MgO, then saturates plagioclase at 5.3 wt% MgO, and magnetite at 4.4 wt% MgO (Fig. 3a-d, A5). The whole rock and melt inclusion data for Guguan from the literature and this study are broadly consistent with this model. Some melt inclusions from this study and those from previous work have higher FeO* concentrations than do whole rock samples from Guguan, suggesting that melt inclusions from Guguan tephras frequently gain Fe. The careful screening procedures in this study eliminate any melt inclusion compositions that deviate significantly from the whole rock compositions, in order to avoid interpreting melt inclusion compositions that have experienced complicated magmatic histories in this work. Another important observation is that the model LLD does not fit the absolute variation between TiO_2 and MgO because the starting composition, chosen as such because of its elevated MgO content, happens to have higher TiO_2 than other whole rock samples from Guguan volcano. Though the line is offset from the data array, the slope of the line however is a good match. In the case of all major elements except for FeO^{actual} and $Fe_2O_3^{actual}$, the fractionation slope defined by the modeled LLD between 5-7 wt% MgO was used to correct for the effects of fractional crystallization (Electronic Appendix E). For FeO^{actual} and $Fe_2O_3^{actual}$, an empirical fractionation slope constrained by the trend in the melt inclusion data from this study was used (Electronic Appendix E).

Sarigan

Tephra sample Sari15-04 is brown in color and show signs of slight alteration. The largest size fraction observed in this tephra sample is ~0.25 cm in diameter. This tephra sample contains crystals of olivine, clinopyroxene, plagioclase, and magnetite.

The modeled LLD begins with sample Sari15-04-27 as the parental composition. Starting at 1.8 kbar and decompressing at a constant rate of 0.9 bar/ºC, this model crystallizes olivine, then saturates clinopyroxene at 7.1 wt% MgO, then saturates magnetite at 6.8 wt% MgO, and then saturates plagioclase at 4.1 wt% MgO (Fig. 3a-d, A6). The whole rock and melt inclusion data for Sarigan from the literature and this study are broadly consistent with this model, with a few exceptions. While the model LLD is a good match for the variation in FeO* and TiO_2 with decreasing MgO in the compositional range of the melt inclusions in this study, whole rock compositions that extend to lower MgO concentrations are not well fit by the model. This may be because magnetite does not saturate in this model and the fractionation of plagioclase drives FeO* and TiO_2 contents to increasingly higher values, opposite to the trends observed in the natural whole rock data. If magnetite is allowed to saturate in the model, it saturates too early and the LLD is inconsistent with the natural data in the compositional range of melt inclusions in this study. Because none of our Sarigan melt inclusions have MgO < 5.0 wt%, we chose the model that better describes the compositional range of samples at higher MgO concentrations. For melt inclusion compositions in this study, in the case of all major elements except for FeO^{actual} and $Fe_2O_3^{actual}$, the fractionation slope defined by the modeled LLD between 5-7 wt% MgO was used to correct for the affects of fractional crystallization (Electronic Appendix E). For FeO^{actual} and $Fe_2O_3^{actual}$, an empirical

fractionation slope constrained by the trend in the melt inclusion data from this study was used (Electronic Appendix E).

NW Rota-1

Submarine pillows from NW Rota-1 are vesicular and have glassy pillow rinds. The pillow interiors have crystals of plagioclase, clinopyroxene, and olivine.

The modeled LLD begins with sample HD488-R11 as the parental composition. This model crystallizes olivine isobarically at 0.1 kbar, then saturates plagioclase at 7.2 wt% MgO, and then saturates clinopyroxene at 6.1 wt% MgO (Fig. 3a-d, A7). Though there is a paucity of data for NW Rota-1, the submarine glass data from this study are broadly consistent with this model and in the case of all major elements except for FeO^{actual} and $Fe_2O_3^{actual}$, the fractionation slope defined by the modeled LLD between 5-7 wt% MgO was used to correct for the effects of fractional crystallization (Electronic Appendix E). For FeO^{actual} and $Fe_2O_3^{actual}$, an empirical fractionation slope constrained by the trend in the submarine glass data from this study was used (Electronic Appendix E).

Northern and Southern Mariana Trough

Northern Mariana trough glasses discussed here were provided only as chipped glass fragments. These glass fragments are fresh and some are vesicular. Southern Mariana trough pillow basalts are vesicular and have glassy pillow rinds. The interior of some vesicle walls are lined with small sulfide crystals. The pillow interiors have olivine ± plagioclase ± clinopyroxene.

Northern Mariana trough samples range in H_2O contents, from 0.5 to 2.5 wt% (Fig. 4d). Water effects magmatic differentiation by suppressing the saturation of plagioclase and clinopyroxene and parental magmas with the same major element

composition but different H_2O concentrations may have different model LLDs (e.g., Lytle *et al.*, 2013). However, the major element variations in the Mariana trough samples are similar and so will be described by a single LLD. The modeled LLD begins with sample 80-1-3 from Stolper and Newman (1994) as the parental composition. This model crystallizes olivine isobarically at 1.5 kbar, then saturates plagioclase at 8.5 wt% MgO, then saturates clinopyroxene at approximately 6.1 wt% MgO, then saturates magnetite at 3.9 wt% MgO (Fig. 3e-h). The submarine glass data from this study are broadly consistent with this model and in the case of all major elements except for FeO^{actual} and $Fe_2O_3^{actual}$, the fractionation slope defined by the modeled LLD between 5-7 wt% MgO was used to correct for the effects of fractional crystallization (Electronic Appendix E). For FeO^{actual} and $Fe_2O_3^{actual}$, an empirical fractionation slope constrained by the trend in the submarine glass data from this study was used (Electronic Appendix E).

Diffusive re-equilibration of melt inclusions

To assess whether melt inclusion suites in this study are significantly changed by diffusive re-equilibration, we compare the trace element variability recorded in suites of melt inclusions from Sarigan, Agrigan, and Alamagan volcanoes to submarine glasses from the southern Mariana trough (Fig. A10; i.e., Cottrell *et al*., 2002, Kent 2008). There is a simple relationship between the variability in trace element compositions within a population of basaltic glasses and the bulk partition coefficient of the element (peridotite/melt); more incompatible elements show greater dynamic range in concentration (Hoffman, 1988). The same correlations should be recorded in suites of melt inclusions if diffusive re-equilibration has not significantly modified the trace

element compositions of the population. To apply this principle to subduction related magmas, we look only at elements that are not recycled from the subducting slab in the Marianas (Fig. A10, Pearce *et al*., 2005) because addition of an element to the mantle wedge by an aqueous fluid, for example, will mask relationships between the element's abundance and variability and the partitioning behavior of the element during simple mantle melting. As expected, there is a correlation between bulk partition coefficient and the percent standard deviation of trace element concentrations measured in southern Mariana trough glasses (Fig. A10, R^2=0.81). Melt inclusion suites from Sarigan and Alamagan are also correlated (Fig. A10, R^2=0.77 and 0.70, respectively), suggesting that their trace element concentrations are not significantly affected by diffusive re-equilibration on the timescales relevant to trace element diffusion in olivine. Melt inclusions from Agrigan volcano, however, do not show this correlation and may thus have experienced some amount of diffusive equilibration (Fig. A10).

We also examine the relationship between the sizes of melt inclusion and their major element composition and Fe redox, to test whether smaller diameter melt inclusions have re-equilibrated and larger diameter melt inclusions have not. There is no relationship between the size of melt inclusion and the major element composition, measured $Fe^{3+}/\sum Fe$ ratios in the inclusions, or the amount of disequilibrium between melt inclusion and olivine host (Fig. A11). The exception to this is the apparent disequilibrium between four Agrigan melt inclusions and their olivine hosts, where the smallest diameter melt inclusions appear to have undergone more post-entrapment crystallization than larger inclusions. These melt inclusions require more than 2% post-entrapment crystallization correction to reach equilibrium with its olivine host and are not considered

in this study. These results suggest either that diffusive re-equilibration (a) has not occurred in melt inclusions used in this study, or (b) has gone to completion such that even slow diffusing elements in olivine (e.g., Ca) have completely re-equilibrated in melt inclusions with 50-300 μm diameter.

Trace element heterogeneity and magma mixing

Melt inclusions and submarine glasses from a single arc volcano commonly record significant heterogeneity in trace element compositions that potentially reflect contributions and mixing between several parental magmas (e.g., Figure 9 in the main text, Fig. A13). If the parent magmas are also heterogeneous in their major element compositions, major element relationships could reflect magma mixing rather than crystal fractionation and this would introduce error into our method for calculating the fO_2 of primary magmas in the Marianas. Figure A13 shows the La/Yb ratios of melt inclusions and submarine glasses versus the Fo# of olivine host (for melt inclusions), SiO_2 content, measured $Fe^{3+}/\sum Fe$ ratios, Na_2O content, TiO_2 content, and K_2O content. The variation in La/Yb ratio versus the Fo# of the olivine host demonstrates the heterogeneity in trace element composition of melt inclusions in this study (vertical dashed line), which is greater than can be expected by simple fractionation of La from Yb during differentiation. Neither La nor Yb are thought to be a significant component slab fluids or sediment melts in the Marianas, and the variation observed here likely reflects heterogeneity in the mantle source and/or variations in the extent of melting at each location. The wide variation in La/Yb ratios (1 to >4) observed within a single suite of melt inclusions occurs in a relatively narrow range of SiO_2 concentrations (44-49 wt%).

There is no correlation between SiO_2, Na_2O, and TiO_2 concentrations, or Fe redox ratios and La/Yb ratios, suggesting that magma is not evident in the major element compositions of the melt inclusions. The exception to this is K_2O concentrations in melt inclusions from Sarigan volcano, in which four melt inclusions with high K_2O concentrations (>0.30) also have La/Yb ratios that are a factor of two higher than the main melt inclusion population (>2.0). However, the high K_2O concentrations of these samples did not influence the choice of the fractionation model used for calculating primary melt compositions. Instead, these inclusions were corrected back to MgO = 7.0 wt% along a shallow slope that fit the main melt inclusion population best, creating a range in primary melt compositions used to calculate primary fO_2. We note that these inclusions cannot be readily distinguished from the main melt inclusion population in other major elements, and that changing the K_2O concentrations of primary melts by a factor of two does not impact calculated fO_2. Additionally, the same trace element heterogeneity recorded by melt inclusion populations is also recorded in submarine glasses from the Mariana trough, Pagan volcano, and NW Rota-1 volcanoes, suggesting that this observation is not simply a melt inclusion phenomenon. We conclude that the major element relationships observed in Figure 3 in the main text, as well as in Figures A2-A7 in the electronic appendix can be reasonably described by simple crystal fractionation.

Sensitivity test of primary melt model results

The method used to calculate primary melt compositions and fO_2 has many sources of error, uncertainty in the inputs, and calculation constraints. Here, we present the results of sensitivity tests of the model results for a subset of samples.

The calculations in the main text reference all primary melt compositions to a mantle olivine at Fo90. If the mantle is more depleted by melt extraction, as might be the case under the volcanic arc, or more fertile, the final endpoint of the calculation may reasonably be Fo91 or Fo89, respectively. Figure A12, panel a, demonstrates the sensitivity of the final calculation to mantle olivine Fo#. The difference between primary fO_2 calculated for arc and back-arc primary melts in equilibrium with Fo89 and Fo90 olivine, or Fo90 and Fo91 olivine, is equal to ~0.07 log units relative to the QFM oxygen buffer. This is an order of magnitude smaller than the observed offset between MORB, Mariana trough, and Mariana arc magmas.

The calculations in the main text correct the major element compositions of samples with MgO between 5 and 7 wt% to a reference MgO = 7.0 wt%, after which the calculation assumes that olivine is the only liquidus phase back to a melt composition in equilibrium with Fo90 olivine. For the arc samples, it is possible that clinopyroxene remains a liquidus phase to higher MgO concentrations. Figure A12, panel b shows the sensitivity of the final calculation to (1) the choice of MgO reference value (blue stars use a reference MgO = 8.0 wt%) and (2) the inclusion of clinopyroxene as a liquidus phase back to equilibrium with Fo90 olivine (pink stars include olivine and clinopyroxene as liquidus phases back to equilibrium with Fo90 olivine). Increasing the MgO reference value to 8.0 wt% increases the primary melt fO_2 by 0.17 log units. If clinopyroxene is included along with olivine as a liquidus

phase, primary melt fO_2 is reduced by 0.23-0.42 log units. Each of these outcomes is less than the difference of primary melt fO_2 between MORB, the Mariana trough, and the Mariana arc.

Finally, there are several model inputs that have some uncertainty associated with them. To assess the total impact of these uncertainties, we have performed a Monte Carlo error analysis. The error analysis uses an uncertainty of 0.5%, 1%, 3%, 2%, and 5% on the raw concentrations of Al_2O_3, FeO*, CaO, Na_2O, K_2O in the glasses associated with electron probe micro analysis and an uncertainty of 3% on the determination of $Fe^{3+}/\sum Fe$ ratios. It uses a 10% uncertainty in the slope of the lines used to calculate the reference compositions for these same elements to MgO7.0. We assign a 20% uncertainty to the constraints on primary melt T and P and a 15% uncertainty in the raw concentrations of Ba and La measured in the glasses by LA-ICPMS. The error simulation for calculating primary melt fO_2 was run through 100 iterations, allowing each variable to vary randomly within the limits of uncertainty described here. The 95 intermediate solutions are shown as error ellipses around the model calculations presented in the main text and shown in Figure A12, panel c. The error ellipses are much smaller than the variation between MORB, Mariana trough and Mariana arc constraints in either Ba/La ratio or primary melt fO_2, indicating that the model results for each point are distinct.

Figure Captions

Figure A1: Example Fe-μ-XANES spectra for San Carlos olivine (green line), standard glass LW+1.0 (brown line; Cottrell *et al.*, 2009), melt inclusion sample Ala03-01 with olivine interference (solid black line) and melt inclusion sample Ala03-01 without olivine

interference (dashed black line). Inset shows the pre-edge feature for all spectra in greater detail. All spectra are normalized to an arbitrary absorption intensity of 1.0 for the region between 7205 and 7210 eV.

Figure A2: Major element variations (FeO*, CaO/Al_2O_3, Al_2O_3, and TiO_2) vs. MgO for Agrigan melt inclusions from tephras Agri04, Agri05, and Agri07, compared to the available literature data for Agrigan melt inclusions and lavas. Large diamonds are melt inclusions used in this study. Small diamonds are olivine hosted melt inclusions that have been eliminated from this study due to Fo_{host}-$Fo_{melt\ inclusion}$ disequilibrium. The light gray diamonds, dotted diamonds, and open diamonds are olivine hosted melt inclusions from Kelley and Cottrell (2012), Kelley *et al.*, (2010), and Shaw *et al.* (2008), respectively. The white circles are whole rock data for tephras collected at Agrigan volcano (Plank, unpublished data). Black circles are whole rock data for lavas collected at Agrigan volcano, compiled using GeoROC (Electronic appendix J). The black line is the Petrolog3 fractionation model, using a starting composition equal to Agri04-05 and fractionating olivine, clinopyroxene, and plagioclase at 1 kbar, using mineral melt models of Roeder and Emslie (1970) and Danyushevsky (2001), treating Fe^{2+}/Fe^{3+} as a closed system (Danyushevsky & Plechov, 2011).

Figure A3: Major element variations (FeO*, CaO/Al_2O_3, Al_2O_3, and TiO_2) vs. MgO for Pagan melt inclusions from tephra Paga8 and submarine glasses, compared to the available literature data for Pagan melt inclusions and lavas. Large squares are a melt inclusion (solid blue) and submarine glasses (checkered) used in this study. The dotted

squares and open squares are olivine hosted melt inclusions from Kelley *et al.* (2010) Shaw *et al.* (2008), respectively. Black circles are whole rock data for lavas collected at Pagan volcano, compiled using GeoROC (Electronic appendix J). The black line is the Petrolog3 fractionation model, using a starting composition equal to HDP1147-R06 and fractionating olivine, plagioclase, clinopyroxene, and magnetite at 3 kbar and decompressing at a rate of 5 bar/°C. Mineral melt models of Roeder and Emslie (1970), Danyushevsky (2001), and Ariskin and Barmina (1999) were used, treating Fe^{2+}/Fe^{3+} as a closed system (Danyushevsky & Plechov, 2011).

Figure A4: Major element variations (FeO*, CaO/Al_2O_3, Al_2O_3, and TiO_2) vs. MgO for Alamagan melt inclusions from tephras Ala02 and Ala03, compared to the available literature data for Alamagan melt inclusions and lavas. Large circles are melt inclusions used in this study. Small circles are olivine hosted melt inclusions that have been eliminated from this study due to Fo_{host}-$Fo_{melt\ inclusion}$ disequilibrium (light green) and to elevated FeO* relative to the literature data (dark green). The open, thick lined circles are olivine hosted melt inclusions from Shaw *et al.* (2008). The open, thin lined circles are whole rock data for tephras collected at Alamagan volcano (Plank, unpublished data). Black circles are whole rock data for lavas collected at Alamagan volcano, compiled using GeoROC (Electronic appendix J). The black line is the Petrolog3 fractionation model, using a starting composition equal to Ala02-01 and fractionating olivine, plagioclase, and clinopyroxene at 2 kbar and decompressing at a rate of 15 bar/°C. Mineral melt models of Roeder and Emslie (1970) and Danyushevsky (2001) were used, treating Fe^{2+}/Fe^{3+} as a closed system (Danyushevsky & Plechov, 2011).

Figure A5: Major element variations (FeO*, CaO/Al_2O_3, Al_2O_3, and TiO_2) vs. MgO for Guguan melt inclusions from tephras Gug11 and Gug23-02, compared to the available literature data for Guguan melt inclusions and lavas. Large hexagons are melt inclusions used in this study. Small hexagons are olivine hosted melt inclusions that have been eliminated from this study due to Fo_{host}-Fo_{melt} inclusion disequilibrium (solid and checkered light yellow). The dotted hexagons, slashed hexagons, and open hexagons are olivine hosted melt inclusions from Kelley *et al.* (2010), Kelley and Cottrell (2009), and Shaw *et al.* (2008), respectively. The white circles are whole rock data for tephras collected at Guguan volcano (Plank, unpublished data). Black circles are whole rock data for lavas collected at Guguan volcano, compiled using GeoROC (Electronic appendix J). The black line is the Petrolog3 fractionation model, using a starting composition equal to SD46-1-1 and fractionating olivine, plagioclase, clinopyroxene, and magnetite at 2 kbar and decompressing at a rate of 5 bar/°C. Mineral melt models of Roeder and Emslie (1970), Danyushevsky (2001), and Ariskin and Barmina (1999) were used, treating Fe^{2+}/Fe^{3+} as a closed system (Danyushevsky & Plechov, 2011).

Figure A6: Major element variations (FeO*, CaO/Al_2O_3, Al_2O_3, and TiO_2) vs. MgO for Sarigan melt inclusions from tephra Sari15-04, compared to the available literature data for Sarigan melt inclusions and lavas. Large stars are melt inclusions used in this study. Small stars are olivine hosted melt inclusions that have been eliminated from this study due to Fo_{host}-Fo_{melt} inclusion disequilibrium (light orange) and to elevated FeO* relative to the literature data (dark orange). The dotted stars and open stars are olivine hosted melt

inclusions from Kelley *et al.* (2010) and Shaw *et al.* (2008), respectively. The white circles are whole rock data for tephras collected at Alamagan volcano (Plank, unpublished data). Black circles are whole rock data for lavas collected at Sarigan volcano, compiled using GeoROC (Electronic appendix J). The black line is the Petrolog3 fractionation model, using a starting composition equal to Sari15-04-27 and fractionating olivine, plagioclase, and clinopyroxene at 1.8 kbar and decompressing at a rate of 9 bar/°C. Mineral melt models of Roeder and Emslie (1970), Danyushevsky (2001), and (Nielsen, 1988) were used, treating Fe^{2+}/Fe^{3+} as a closed system (Danyushevsky & Plechov, 2011).

Figure A7: Major element variations (FeO*, CaO/Al_2O_3, Al_2O_3, and TiO_2) vs. MgO for NW Rota-1 submarine glasses. The black line is the Petrolog3 fractionation model, using a starting composition equal to HD288-R11 and fractionating olivine, plagioclase, and clinopyroxene at 0.1 kbar. Mineral melt models of Roeder and Emslie (1970), and Danyushevsky (2001) were used, treating Fe^{2+}/Fe^{3+} as a closed system (Danyushevsky & Plechov, 2011).

Figure A8: Calculated Fo# of an equilibrium olivine host for each melt inclusion included in this study, using an FeO/MgO $K_D = 0.3$ (Roeder & Emslie, 1970) vs. the measured Fo# of the olivine host. Solid black line is a 1:1 relationship. Dashed black lines represent the error envelope of the equilibrium Fo# calculation. This error envelope is calculated by propagating average analytical error for FeO* and MgO through the equations for

calculating equilibrium Fo#. Any melt inclusion that falls below the 1:1 line is subject to the PEC correction described in the main text, until the melt inclusion composition is in equilibrium with the composition of its olivine host. Any melt inclusion composition that falls above the 1:1 line, but lies within the error envelope, is left uncorrected.

Figure A9: Plots of V vs. MgO variation for Mariana trough samples (southern Mariana trough, this study; white circles are Mariana trough data from Pearce *et al.*, 2005). The arrow shows the general expectation of the effect of magnetite fractionation on V concentrations in a magma.

Figure A10: A plot of percent standard deviation of select trace element concentrations within a population of samples versus bulk $D^{perid/melt}$ for melt inclusions from Sarigan, Agrigan, and Alamagan volcanoes, as well as submarine glasses from the southern Mariana trough. Bulk $D^{perid/melt}$ are from Cottrell *et al.*, (2002).

Figure A11: Plots of the size of melt inclusions versus CaO, $Fe^{3+}/\Sigma Fe$ ratios, and the apparent disequilibrium between melt inclusion and olivine host pairs. Melt inclusion shapes vary between circular and oblate. In the cases where the melt inclusions are oblate, the left hand panels use the short axis and the right hand panels use the long axis of the inclusions. In the bottom panels, the gray dashed line marks the position of Fo_{EQ}-Fo_{host} equal to 0, or apparent equilibrium between melt inclusion and olivine host. Any melt inclusion within the gray box are those that are used in this study.

Figure A12: Plots demonstrating the results of sensitivity tests for the calculations in this study (panels a and b) and a Monte Carlo error simulation demonstrating the effects of accumulated errors on the final calculation (panel c).

Figure A13: Plots of La/Yb ratios in melt inclusions and submarine glasses versus (a) Fo# of the olivine host, (b) SiO_2, (c) $Fe^{3+}/\sum Fe$ ratios, (d) Na_2O, (e) TiO_2, and (f) K_2O. The vertical dashed line emphasizes the heterogeneity in La/Yb ratio at fixed values for Fo#, SiO_2, or $Fe^{3+}/\sum Fe$ ratio that is unrelated to crystal fractionation. The solid line illustrates the direction that crystal fractionation drives the variables.

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